NOx Control with Ammonia-Free SCR

Zhen Fan
Foster Wheeler North America Corp, 12 Peach Tree Hill Road, Livingston, NJ 07039
zhen_fan@fwc.com, Tel: 973-535-2548, Fax: 973-535-2242

Andrew H. Seltzer
Foster Wheeler North America Corp, 12 Peach Tree Hill Road, Livingston, NJ 07039
Andrew_seltzer@fwc.com, Tel: 973-535-2542; Fax: 973-535-2242

Richard G. Herman
Energy Research Center, Lehigh University, 117 Atlss Drive, Bethlehem, PA 18015
rgh1@lehigh.edu, Tel: 610-758-3486; Fax: 610-758-6536

Kamalendu Das
NETL, 3610 Collins Ferry Road, P.O. Box 880, Morgantown, WV 26507
kamalendu.das@netl.doe.gov, Tel: 304-285-4065

Summary

This paper introduces a novel ammonia-free NOx control system, where the process uses natural CO from the boiler as reductant for NOx reduction. It optimizes burner and furnace to achieve a low level of NOx from the boiler and to provide an adequate amount of CO for NOx reduction over a base-metal based catalyst downstream. In this catalyzed redox process, the excess CO, after being used as a NOx reductant, is efficiently oxidized to CO_2 by O_2 in the flue gas on the same catalyst. The research work presented in this paper is part of a novel multi-emission control system proposed utilizing injected catalyzed activated carbon as a catalyst and a sorbent, where low cost base-metals are applied as active components for the catalyst. In this system, in addition to ammonia-free NOx reduction and CO oxidation, the catalyst also functions as a sorbent to absorb SO_2. This SOx adsorbed catalyst is then applied as sorbent to capture mercury from flue gas.

This paper concentrates on the furnace optimization work simulated with CFD model to suppress NOx and to provide CO, on the experimental work performed with a bench scale reactor to evaluate performance of different catalysts, and on the design work of scaling-up for the overall process.

This new concept breaks the constraining relationship between furnace NOx and CO emissions. By modifying furnace operating conditions only, such as by reducing the overall excess air level, and/or by increasing over-fire air without physical changes to the burner and boiler equipment, furnace outlet NOx can be reduced from 250-400 ppm to about 150 ppm, and adequate CO/NO ratio can be obtained. The CFD study predicts with these adjustments that the effects on boiler performance such as unburned carbon, boiler efficiency, and furnace exit gas temperature, are relatively small and within the normal range seen in power plant operations. Trading off NOx by CO in the furnace brings potential reduction of downstream SCR duty, size and ammonia requirement.

To support the NOx reduction by CO on catalyst, base metal (Cu, Fe, K, and Ce) based catalyst on activated carbon (AC) and activated alumina (AA) prepared and tested in this study exhibited very promising NOx reduction under simulated flue gas conditions including 3% oxygen. NOx reduction from 260 ppm to 25-50 ppm was achieved with operating temperatures ranging from 230-350^{o}C. In combining with boiler NOx reduction from furnace adjustment, a total over 90% of NOx reduction can be obtained. All tested catalysts showed high selectivity toward N_2 during NOx reduction. The catalyst also oxidized over 80-90% of the CO in the gas in a wide temperature range from 140-350^{o}C. It has been found that the conversions of CO to CO_2 on the catalyst were always high even at low temperature regardless of NOx conversion. The catalyst can be used as a polishing stage for emission of CO to allow the boiler system to be optimized for NOx reduction. In consideration of competition reaction between reductant of CO and carbon, and between oxidant of NOx and O_2, the catalyst has been so designed that it performed better selections to enhance both NOx reduction and CO depletion, and to suppress side reactions. Only some of the tested catalysts performed well in a wide temperature range, and some of them promoted CO generation from partial combustion of activated carbon as temperature increased, and showed runaway of CO without enough depletion. From test, a high gas hourly space velocity (GHSV) shifted reaction temperature high, which resulted in changes of reaction pathway and selectivity for some cases. The dependence of reactivity of catalysts on active components, component ratios, and component loadings, as well as operation temperatures has been studied. Based on these studies, a method to enhance NOx reduction reactivity of the catalyst has been found.

The solid catalyst can be applied in various configurations, such as fixed bed, fluidized bed, and entrained flow reactor, depending on the process requirement and the performance of the catalyst, as well as the cost of operation. In test catalysts functioned differently in suffering from sulfur poison to the extent of fast, slow and minor deactivations. For the injection option, due to short residence time, the initial high reactivity is important but not the deactivation. On the other hand, for the fixed bed option, long life of the catalyst becomes very important. The tests of catalysts corresponded to the different purposes. Detailed test data analysis and catalyst reaction modeling indicate that the effectiveness of the catalyst is close to 1.0 for the tested samples and conditions without significant limits from bulk mass transfer and pore diffusion. Preliminary scale-up correlations have been obtained for commercial design.

In summary, a new approach to control NOx and CO emissions by ammonia-free SCR is proposed, where instead of ammonia, boiler natural CO is used as reductant. The development of a catalyst has been proceeding with promising results. The financial support of this study by the U.S. DOE NETL is appreciatively acknowledged.